

PCT

WORLD INTELLECTUAL PROPERTY ORGANIZATION  
International Bureau



INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

(51) International Patent Classification 6 : <b>B32B 27/08</b>	<b>A1</b>	(11) International Publication Number: <b>WO 97/01438</b> (43) International Publication Date: <b>16 January 1997 (16.01.97)</b>
-------------------------------------------------------------------	-----------	-------------------------------------------------------------------------------------------------------------------------------------

(21) International Application Number: <b>PCT/US95/08164</b> (22) International Filing Date: <b>28 June 1995 (28.06.95)</b>	(81) Designated States: AU, CA, CN, CZ, HU, JP, KR, NZ, PL, SK, European patent (AT, BE, CH, DE, DK, ES, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE).
(71) Applicant: <b>MOBIL OIL CORPORATION [US/US]; 3225 Gallows Road, Fairfax, VA 22037-0001 (US).</b>  (72) Inventors: <b>BENOIT, Gordon, Leon; 49 Latchmere Drive, Victor, NY 14564-1282 (US). VANDERVELDEN, Rudolf; 250 Hook Road, Macedon, NY 14502-9612 (US).</b>  (74) Agents: <b>SUNG, Tak, K. et al.; Mobil Oil Corporation, 3225 Gallows Road, Fairfax, VA 22037-0001 (US).</b>	Published <i>With international search report.</i>

(54) Title: **MULTILAYER FILM STRUCTURES FOR USE IN THE PRODUCTION OF BANKNOTES OR THE LIKE**

(57) Abstract

A laminate multilayer film substrate for use in the production of banknotes including a first layer and a second layer, which may be the same or different, each comprising at least 50 weight percent of a high density polyethylene having a density of at least .94 g/cc and each being oriented in at least a first direction to a degree which is at least three times greater than the degree of orientation present in a second direction substantially normal to the first direction wherein the second layer is laminated to the first layer so that the first direction of orientation of the second layer is substantially aligned with the first direction of orientation of the first layer.

**FOR THE PURPOSES OF INFORMATION ONLY**

Codes used to identify States party to the PCT on the front pages of pamphlets publishing international applications under the PCT.

AM	Armenia	GB	United Kingdom	MW	Malawi
AT	Austria	GE	Georgia	MX	Mexico
AU	Australia	GN	Guinea	NE	Niger
BB	Barbados	GR	Greece	NL	Netherlands
BE	Belgium	HU	Hungary	NO	Norway
BF	Burkina Faso	IE	Ireland	NZ	New Zealand
BG	Bulgaria	IT	Italy	PL	Poland
BJ	Benin	JP	Japan	PT	Portugal
BR	Brazil	KE	Kenya	RO	Romania
BY	Belarus	KG	Kyrgyzstan	RU	Russian Federation
CA	Canada	KP	Democratic People's Republic of Korea	SD	Sudan
CF	Central African Republic	KR	Republic of Korea	SE	Sweden
CG	Congo	KZ	Kazakhstan	SG	Singapore
CH	Switzerland	LI	Liechtenstein	SI	Slovenia
CI	Côte d'Ivoire	LK	Sri Lanka	SK	Slovakia
CM	Cameroon	LR	Liberia	SN	Senegal
CN	China	LT	Lithuania	SZ	Swaziland
CS	Czechoslovakia	LU	Luxembourg	TD	Chad
CZ	Czech Republic	LV	Latvia	TG	Togo
DE	Germany	MC	Monaco	TJ	Tajikistan
DK	Denmark	MD	Republic of Moldova	TT	Trinidad and Tobago
EE	Estonia	MG	Madagascar	UA	Ukraine
ES	Spain	ML	Mali	UG	Uganda
FI	Finland	MN	Mongolia	US	United States of America
FR	France	MR	Mauritania	UZ	Uzbekistan
GA	Gabon			VN	Viet Nam

- 1 -

MULTILAYER FILM STRUCTURES FOR USE IN THE  
PRODUCTION OF BANKNOTES OR THE LIKE

The present invention relates to a multilayer film for use in the production of paper-like products such as bank notes, 5 security documents, including travellers and bank checks, and to a process for their production. More particularly, the present invention relates to a laminated multilayer film structure having the characteristics of the high quality papers typically employed in the production of bank notes and security 10 documents.

In the production of bank notes and security documents rag paper has been employed for over 300 years. As is well known, rag paper has several properties which are highly desirable in such applications, including dead foldability, tear resistance, 15 printability and embossability.

These highly desired properties may be characterized as follows: deadfold is the ability of a substrate to be creased or folded and to retain the fold without opening. Tear resistance is the ability of a substrate to resist both 20 initiated and uninitiated tears and punctures. Printability is the ability of the substrate to adsorb and bond inks used during the lithographic printing process. Embossability is the ability of the substrate to deform under the pressures of the intaglio printing process to form a raised image on the resulting bank 25 note or security document, with the intaglio ink remaining on the raised, deformed region resulting in a high degree of tactility or feel to the bank note or security document. As may be appreciated, these properties combine to give bank notes and the like their familiar feel and functionality.

30 With the advent of color copiers and computer graphic scanners, the counterfeiting of bank notes has markedly increased. While there are active programs underway by major currency paper producers to make their substrate more secure through the use of watermarks, metallized threads and optical 35 variable devices (OVDs), such as photochromics, holographics, and diffraction gratings, these efforts do not appear to hold much promise of thwarting counterfeiters.

- 2 -

Plastic substrates offer a major security feature if a clear "window" is incorporated into the bank note. This window would ensure that a scanner or color copier could not copy the note. Additionally, other security features can be incorporated 5 into or onto the bank note, including reverse printing of the note to protect the security devices and the print.

In accordance with the present invention, there is provided a curl-resistant, cross-sectionally symmetrical, laminated multilayer film substrate for use in the production of 10 banknotes, security papers comprising:

(a) a first layer having inner and outer sides, comprising at least 50 weight percent of a high density polyethylene having a density of at least 0.94, said first layer being oriented in at least a first direction to a degree which is at least three 15 times greater than the degree of orientation present in a second direction substantially normal to the first direction; and

(b) a second layer having inner and outer sides, comprising at least 50 weight percent of a high density polyethylene having a density of at least 0.94, said second layer being oriented in 20 at least a first direction to a degree which is at least three times greater than the degree of orientation present in a second direction substantially normal to the first direction;

(c) a laminating adhesive resin disposed between the inner sides of (a) and (b), which comprises a component selected from 25 the group consisting of low density polyethylene (LDPE) and linear low density polyethylene (LLDPE); said second layer being laminated to said first layer so that the first direction of orientation of said second layer is substantially aligned with the first direction of orientation of said first layer.

30 In another embodiment, the present invention relates to a method for preparing a bank note which comprises:

i) laminating (a) a first layer having inner and outer sides, comprising at least 50 weight percent of a high density polyethylene having a density of at least 0.94 g/cc, 35 said first layer being oriented in the transverse direction (TD) to a degree which is at least three times greater than the degree of orientation present in the machine direction (MD) to

- 3 -

(b) a second layer having inner and outer sides, comprising at least 50 weight percent of a high density polyethylene having a density of at least 0.94, said second layer being oriented in the transverse direction to a degree which is at least three times greater than the degree of orientation present in the machine direction, with a laminating adhesive resin which comprises a component selected from the group consisting of low density polyethylene (LDPE) and linear low density polyethylene (LLDPE), said second layer being laminated in the film substrate so that said transverse direction of orientation of said second layer is substantially aligned with said transverse direction of orientation of said first layer, thereby forming an embossable substrate; and

ii) embossing said substrate.

15 The resultant films exhibit good dead-fold characteristics and other properties, including a symmetrical structure which provides curl free film even at elevated temperatures, e.g., 66°C (150°F), making them highly suited for the production of bank notes and other security documents.

20 In still another embodiment, the present invention relates to a security token such as a banknote or stock certificate prepared from an embossable, laminated multilayer film substrate, said substrate comprising:

(a) a first layer having inner and outer sides, comprising at least 50 weight percent of a high density polyethylene having a density of at least 0.94, said first layer being oriented in the transverse direction (TD) to a degree which is at least three times greater than the degree of orientation present in the machine direction (MD); and

30 (b) a second layer having inner and outer sides, comprising at least 50 weight percent of a high density polyethylene having a density of at least 0.94, said second layer being oriented in the transverse direction to a degree which is at least three times greater than the degree of orientation present in the machine direction, said second layer being laminated to said first layer so that the transverse direction of orientation of said second layer is substantially aligned with the transverse

- 4 -

direction of orientation of said first layer.

In still another embodiment, the present invention relates to a method for preparing an embossed, curl-resistant laminated multilayer film substrate for use in the production of 5 banknotes, security papers and the like which comprises:

i) providing copolymer polypropylene skins on both sides of an oriented high density polyethylene layer having inner and outer sides and comprising at least 50 weight percent of a high density polyethylene having a density of at least 10 0.94, said layer being oriented in the transverse direction (TD) to a degree which is at least three times greater than the degree of orientation present in the machine direction (MD);

ii) coating said copolymer propylene skin provided on one side of said layer with an adhesion promoting primer 15 thereby providing a first primed layer (a) having a primed surface;

iii) repeating steps i) and ii) to provide a second primed layer (b) having a primed surface;

iv) providing a discrete security device on said 20 primed surface of one of said primed layers;

v) providing an orientable laminating adhesive resin between said primed layers and laminating said primed layers together while orienting said laminating adhesive resin to provide a substantially transparent laminate;

25 vi) gravure printing at least one side of said substantially transparent laminate with an opacifying ink in register with said security device thereby providing a clear window in register with said security device to render it visible from at least one side, preferably from either side, of 30 said film substrate.

In forming the multilayer film substrates for use in the production of the bank notes and other security documents of the present invention, at least two layers of the substrate are to contain a major proportion of a high density polyethylene (HDPE) 35 having a density of at least 0.94, preferably at least 0.945. These film layers may be composed exclusively of a single HDPE resin, a mixture of HDPE resins, or of HDPE containing a minor

- 5 -

proportion of another polymeric material, such as low density polyethylene (LDPE), linear low density polyethylene (LLDPE), polypropylene, ethylene vinyl alcohol (EVOH) copolymer, ethylene propylene (EP) copolymer or ethylene propylene butene-1 (EPB) 5 copolymer, although a single HDPE resin or a blend of HDPE resins is particularly preferred in the practice of the present invention. The use of processing additives, such as microcrystalline wax or the like may be employed with the preferred HDPE resins to yield improved processing 10 characteristics in the extruder by reducing extruder torque. Films made with either a blend of HDPE resins or with microcrystalline wax have been found to reduce the splittiness of the film which manifests itself as the tendency of the film to break in the TD direction.

15 When blends of HDPE polymers are employed, such blends can comprise two or more polymers all of which preferably have densities of 0.94 or greater. Blends of HDPE polymers advantageously comprise a major proportion of HDPE having a melt index of 0.6 to 1.2 and one or more polymers having a different 20 melt index.

Terblends may also be desirable. Suitable terblends generally comprise 50 to 98 weight percent, preferably 84 to 96 weight percent of HDPE having a density of 0.96 or higher and a melt index of greater than 0.5 to 2.0; 1 to 25 weight percent, 25 preferably 3 to 8 weight percent of HDPE having a density of 0.94 or greater and a melt index of 0.1 to 0.5; and 1 to 25 weight percent, preferably 3 to 8 weight percent, of HDPE having a density of 0.96 or higher and a melt index of greater than 2 to about 8. Preferably, the second and third HDPE polymers 30 which are minor components are present in about equal amounts.

As is particularly preferred, the film substrate of the present invention includes a) a first layer comprising at least 50 weight percent of a high density polyethylene having a density of at least 0.94, the first layer oriented in at least 35 a first direction, e.g., transverse direction (TD), to a degree which is at least three times greater than the degree of orientation present in a direction substantially normal to the

- 6 -

first direction, e.g., machine direction (MD), and b) a second layer also comprising at least 50 weight percent of a high density polyethylene having a density of at least 0.95, the second layer also oriented in at least a first direction, e.g., 5 TD, to a degree which is at least three times greater than the degree of orientation present in a direction, e.g., MD, substantially normal to the first direction, the second layer being laminated to the film substrate so that the first direction of orientation of the second layer is substantially 10 parallel to (or aligned with) the first direction of orientation of the first layer. A method of producing HDPE films with imbalanced biaxial orientation is disclosed in U.S. Patent No. 4,870,122.

As may be appreciated, to achieve the object of improved 15 tear resistance in a multilayer film of the type described herein, it has been discovered that imbalanced biaxially oriented HDPE films, laminated so that their primary directions of orientation are aligned substantially parallel to each other, can exhibit increased tear resistance where an orientable 20 laminating resin, e.g., LLDPE, is disposed between the layers and subjected to orienting so that the primary direction of orientation in the oriented resin is substantially normal to the primary direction of orientation of the first and second HDPE film layers. In one embodiment, such orientation in the primary 25 direction of orientation of the laminating adhesive resin is achieved during the laminating step itself, preferably in the machine direction (MD).

The degree of orientation of the HDPE film layers is an important aspect of this invention inasmuch as the proper degree 30 of orientation provides desirable physical properties. Although, higher density HDPE resin having a density of 0.957 or greater can be made directly into thin films by cast extrusion, problems of curling, uniformity and flatness exist. Accordingly, thin HDPE films of 0.02 to 0.04 mm (0.8 to 1.5 mils) having the best 35 balance of properties are obtained using imbalanced biaxially oriented films prepared from films having a cast gauge of 0.3 to 0.5 mm (12 to 20 mils) which are reduced to the desired gauge

- 7 -

by stretching, i.e., orientation.

The films are produced and oriented in a conventional manner. The film is heated to its orientation temperature and first subjected to MD orientation between two sets of nip rolls, 5 the second rotating at a greater speed than the first in an amount equal to the desired draw ratio. Then the film is TD oriented by heating and subjecting it to transverse stretching in a tenter frame. Typically MD orientation is conducted at 60° to 120°C and TD orientation at 110° to 145°C.

10 While it is preferred that the degree of orientation in a first film direction be at least three times greater than the degree of orientation present in a direction substantially normal to the first direction, it is more particularly preferred that each HDPE film layer be oriented to an extent of 1.1 to 2.0 15 times in the machine direction (MD) and 6 to 12 times in the transverse direction (TD). It has been found that the HDPE film layers can be produced with excellent quality at caster speeds of up to 110 fpm corresponding to line speeds of 140 fpm at 1.25 times MD orientation.

20 When employed, this degree of imbalanced orientation produces an interesting effect in the HDPE components of the structure. The effect is a visible rippled and striated appearance, with the ripples being parallel to the transverse orientation direction. Under low magnification, in each square 25 centimeter of HDPE film there will be seen from 5 to 30 discontinuous undulating ripples and striations generally parallel to the direction of orientation. This effect gives the film a slight translucent appearance, which tends to slightly blur distant objects viewed through the film. This effect 30 indicates that the layers have been oriented in an imbalanced manner. The high density polyethylenes contemplated for use in the practice of the present invention include those disclosed in U.S. Patent No. 4,870,122.

To achieve the desired surface characteristics required of 35 the paper-like products of the present invention, one or more skin layers can be applied, in any known manner, to the multilayer HDPE substrate material, for example by coating or

- 8 -

coextrusion before orientation or by coating the HDPE after one or both of the orientation operations. The skin layer can be any of the conventional materials used for this purpose in conjunction with polyolefin films, particularly polyethylene 5 films. For example, to achieve a press-ready surface, a polymeric resin could be blended with fillers, fibers, pigments or the like, as necessary. Additionally, voided films, such as those disclosed in U.S. Patent Nos. 4,377,616, 4,632,869, 4,758,462 and others, may be laminated to the multilayer HDPE 10 substrate to impart the properties of those structures to the films of the present invention.

The HDPE-containing layers (a) and (b) as disclosed above can further comprise copolymer polypropylene skins, e.g., ethylene-propylene-butene-1 terpolymer, provided on at least one 15 side thereof, preferably on both the inner and outer sides thereof. In a particular preferred embodiment, the skins themselves can also comprise a component which is similar to components in the adhesive resin used to laminate layers (a) and (b). For example, in a particularly preferred embodiment, low 20 density polyethylene (LDPE) can make up from 10 to 20 wt% of the skins, with the balance being a copolymer polypropylene.

A suitable adhesion-promoting primer which provides greater adhesion between the laminated surfaces, e.g., polymers selected 25 from the group consisting of polyethyleneimide, epoxy, polyurethane, and acrylic, can be provided between the copolymer polypropylene skins (on the inner sides of (a) and (b)) and the laminating adhesive resin. Primer compositions are disclosed in U.S. Pat. Nos. 4,447,494 and 4,681,803.

It is also envisioned that the substrate can be embossed, 30 dyed, printed, texturised or otherwise treated before or after lamination; this being done on the internal or external surfaces of the laminated layers, so as to provide, for example, visual and/or tactile identification of the nature of a banknote, its significance or value.

35 The laminating techniques which can be employed to effect the present invention are known in the art and include: adhesive-bonding or cementing, e.g., with laminating adhesive

- 9 -

resins, preferably with a transparent agent; solvent-bonding, where a mist of solvent is sprayed over the surfaces to be bonded together; thermal lamination by heat-bonding where thermoplastic sheets are subject to a hot rolling or pressing 5 operation; cast-lamination where one layer is cast onto the second and the second forms a substrate; or, extrusion or draw-lamination as in calendering operations known in the art.

When discrete security devices are incorporated within the substrate, e.g., optically-variable devices (OVDs), they can be 10 enclosed in pouches affixed to the substrate. On the other hand, the optically-variable devices themselves may be incorporated in one (or both) layers of the laminated substrate or between the layers, it not being necessary to incorporate a physically discrete device within a clearly defined pouch formed 15 between the layers.

Any suitable security device can be employed in the present invention such as one selected from the group consisting of optically variable devices (OVDs), magnetic devices, electronic devices, and rare earth element-containing devices, with OVDs 20 particularly preferred.

As employed in the present specification, the term "optically-variable" is used to denote any device which can readily be made to change appearance in a reversible, predictable and reproducible manner. The appearance of such 25 devices may be altered, for example, by the application of body-heat or manual pressure, the variation of the angle of viewing and, the lighting conditions under which viewing takes place. The type of devices envisioned by the present invention are: diffraction gratings, liquid crystals, moire patterns and 30 similar patterns produced by cross-gratings with or without superimposed, refractive, lenticular and transparent grids, such as Fresnel lenses, spaced partially-reflective, and partially transparent, coatings yielding variable interference patterns or the like, bi-refringent or polarizing layers or zone-plates.

35 Generally, optically-active devices of this nature are readily recognized by unskilled persons and are yet extremely difficult to reproduce by photographic and printing techniques.

- 10 -

Moreover the production of any one such device in a reproducible fashion and the incorporation of such a device in a plastic laminate as described by the present invention is likely to be beyond the resources of the great majority of would-be forgers.

5 Where a flexible paper-like product such as a bank-note is sought, it is of course preferable that the optically-variable devices should, themselves, be sheet-like, flexible and thin; it is also preferable for such devices to be compatible with the plastic material employed for the laminae to facilitate bonding

10 and mitigate against reactive changes occurring with time.

According to the present invention, one preferred form of optically variable device may be a reflecting diffraction grating consisting of a metallized thermoplastic film embossed with a diffraction pattern. To prevent access to the embossed 15 pattern for the purpose of illicit replication, it is preferable according to the present invention to employ a layer of thermoplastic material on each side of the metallized film which has similar solubility characteristics to that of the metal layer so that separation by preferential etching will be 20 rendered extremely difficult. Another preferred device is a moire pattern formed by photographically reproducing fine line or dot patterns on each side of a thin film. The spacings of the dots and lines can be readily made too fine to be reproduced by printing techniques and yet the moire pattern can be 25 displayed upon a much larger scale. Unique diffraction and moire patterns will often be preferred for use in bank notes and techniques are available for producing those by computer and photo-reduction methods.

In the production of low denomination bank notes, a 30 suitable level of security against counterfeiting may be obtained by merely providing a clear "window" through the bank note. As indicated above, such a window would ensure that a scanner or color copier could not copy the note. Additionally, other security features can be incorporated into or onto the 35 bank note, including reverse printing of the note to protect the security devices and the print.

It has been found that films similar to those of the

- 11 -

present invention but which are "cross-oriented," can be susceptible to curling at temperatures above 66° (150°F). Such cross-oriented films are similar to those of the present invention except that the second layer is laminated to the film substrate so that the first (primary) direction of orientation of the second layer is substantially normal to the first (primary) direction of orientation of the first layer. It is believed that such curling can result from differences in shrinkage at high temperatures in the machine direction and transverse direction of each layer. Additional curling susceptibility can result where coatings or skins of the layers have different coefficients of contraction from the HDPE component of the layers. Such imbalanced contraction and its attendant curling can be avoided by counterbalancing the overall shrinkage properties of one layer by laminating thereto a second layer identical to the first layer which is provided as a mirror image of the first layer. In other words, one half of the layered film structure is a mirror image of the other, with the plane of symmetry being along the horizontal midline of the layered film structure cross- section. This provides a cross-sectionally symmetrical layered film structure. Examples of such films include those of ABA, ABBA, ABCCBA and ABCDCBA construction where each letter represents a film layer, skin, coating, or adhesive layer. Such a cross-sectionally symmetrical layered film structure is necessarily a "parallel-oriented" structure, i.e., the primary direction of orientation of the first layer is parallel to the primary direction of orientation of the second layer in order to meet the mirror image requirement. Such a construction provides a symmetrical structure wherein the opposing shrinkage forces counteract each other to a significant degree. However, such a two layer parallel construction can be susceptible to poor tear properties in one direction, e.g., TD where two TD oriented films are employed.

It has now been found that using a linear low density polyethylene (LLDPE) laminating resin oriented in the primary direction of orientation which is substantially perpendicular

- 12 -

to the primary direction of orientation of the layers which it ties, evinces reduced susceptibility to tearing. The laminating resin can itself be subjected to orienting to some extent during lamination, preferably after the resin has at least partially 5 or completely solidified, and preferably in the machine direction. Such orienting can be from 1.5 to 10 times, preferably from 4 to 6 times, corresponding to a total drawdown of the laminating adhesive resin of greater than 10%, preferably greater than 20%, say, 75%.

10 It has been found that as molecular weight of the laminating adhesive resin employed is increased, e.g., higher molecular weight LLDPE, orientation effects resulting from the lamination procedure are improved, resulting in a multilayer film of improved tear resistance in both machine direction and 15 transverse direction while exhibiting curl resistance as well.

In one particularly preferred embodiment, the extrusion laminating adhesive resin is itself of ABA construction, i.e., the resin has a core of LLDPE (B layer) and outer or skin layers of LDPE (A layers). The ABA construction for the laminating 20 resin provides an advantage inasmuch the A layers provide a high bond strength between the laminating adhesive resin and the oriented HDPE layers, while the higher molecular weight core (B layer) provides tear resistance in the transverse direction as a result of orienting in the machine direction during 25 lamination. A laminating adhesive resin comprising LDPE only can result in a bond strength ranging from 300 to 800 grams/inch unprimed, and 600 to 1400 g/in with primed HDPE layers. Such bond strengths provide a product which is very difficult to delaminate and which therefore reduces the possibilities of 30 tampering with any security device disposed between the HDPE layers. Use of the ABA laminating adhesive resin provides similar bond strengths while increasing transverse direction tear resistance (g/mil) by a factor of at least two.

The following Examples illustrate the invention in which 35 all parts are by weight unless otherwise specified.

Example 1

This example demonstrates the preparation of a multilayer

- 13 -

film substrate produced in accordance with the present invention which is suitable for preparing banknotes having good dead fold characteristics.

A multilayer layer oriented film substrate having a 1.15 5 mil final thickness was prepared by coextruding HDPE with copolymer polypropylene skins on both sides to form a first layer (a). The HDPE resin employed was Oxychem M-6211, available from Occidental Chemical Corporation, Dallas, Texas, having a density of 0.96 and a melt index of 1.0. The copolymer 10 polypropylene skins comprised 90% Chisso 7510, an ethylene-propylene-butene-1 terpolymer available from Chisso Corporation of Japan, and 10% Mobil LKA-753, a low density polyethylene available from Mobil Chemical Co., Norwalk, Connecticut. The HDPE comprised 90% of the resulting film layer (a) while the 15 skins comprise 10% (5% on each side). A second film layer (b) identical to (a) was also formed. The first and second films (a) and (b) were then oriented 1.4 times in the MD at about 115°C and 6 to 12 times, e.g., 10 times, in the TD direction at 115-140°C in a tenter frame.

20 The copolymer skins on the inner side of oriented layers (a) and (b) were then coated with a polyethylenimine (PEI) primer to form clear 1.15 mil OHD films.

On film (a) can be deposited optical varying devices (OVDs), at predetermined distances from one another so as to 25 provide an OVD at the same location on each of the banknotes which are ultimately prepared from the film.

Films (a) and (b) were laminated by providing therebetween a laminating adhesive LDPE resin, Chevron 1017, available from Chevron Chemical Co., Houston, Texas. The laminating adhesive 30 resin undergoes orienting in the machine direction during the laminating step itself, thereby imparting enhanced tear resistance in the transverse direction (TD). Lamination is carried out by conventional techniques, resulting in a multilayer film substrate having a final thickness of about 3.0 35 mils, the laminating adhesive resin itself providing about 0.7 mils thickness.

The resulting substrate was substantially clear and its

- 14 -

physical properties are summarized in the Table below.

The substrate was then gravure printed on both sides using an opacifying ink. The gravure printing was carried out in such a way that the opacifying ink or coating was applied in register 5 with the OVD embedded within the substrate, i.e., the coating was applied so as to avoid coverage of the OVD, thereby providing a clear window in register with said device, rendering the OVD visible from both sides of the printed substrate. The 10 gravure printed substrate is 3.75 mils in thickness and can be taken up on a roll after printing or sheeted and guillotined in register to provide press-ready sheets of approximately 30 inches by 30 inches which are then packaged in units of 5000 sheets. The product exhibited good dead fold properties, tear 15 resistance in both directions, printfastness, and provided better resolution while requiring less ink during printing than paper bank note substrates. Moreover the product was not susceptible to curling at high storage temperature (180°F for 24 hours). The characteristics of the product thus permit its 20 advantageous substitution for conventional paper substrates used in printing banknotes.

The product is also difficultly delaminatable, rendering isolation of an OVD for counterfeiting purposes extremely difficult, owing to the high bond strengths between the layers of films (a) and (b).

25 Example 2

Example 1 was repeated except that the laminating adhesive resin employed was an LLDPE laminating grade adhesive resin, Dowlex 3010, available from Dow Chemical Co. of Midland, Michigan. The characteristics for the resulting substrate are 30 set out in the Table below. Use of the higher molecular weight LLDPE as the adhesive resin in Example 2 resulted in increased TD tear strength.

Example 3

Example 1 is repeated except that the laminating adhesive 35 resin employed is of ABA construction, A being LDPE, making up 20% of the total laminating resin (10% per side) and B being LLDPE, making up 80% of the total laminating resin. The

- 15 -

characteristics for the resulting substrate are set out in the Table below. Use of the ABA laminating adhesive resin between layers (a) and (b) provides high bond strengths between the layers of films (a) and (b), which is believed attributable to 5 the presence of LDPE components in both the laminating adhesive resin and the copolymer skins which are laminated to one another. Moreover, the presence of more readily orientable higher molecular weight LLDPE in the core of the laminating adhesive resin provides excellent TD tear strength resulting 10 from orienting in the machine direction of the ABA laminating adhesive resin.

TABLE

		<u>Ex. 1</u>	<u>Ex. 2</u>	<u>Ex. 3</u>
15	Tensile (psi x 10 <sup>3</sup> )	MD MD	6 16	6 20
	Modulus	MD	365	380
	Stiffness (grams)	MD TD	17 21	20 24
20	Tear Strength (g/mil)	MD TD	89 4	63 5
				20

CLAIMS

1. A laminated multilayer film substrate which comprises a first layer (a) and a second layer (b), which may be the same 5 or different, each comprising at least 50 weight percent of a high density polyethylene having a density of at least 0.94, and each being oriented in at least a first direction to a degree which is at least three times greater than the degree of orientation present in a second direction substantially normal 10 to the first direction; and (b) being laminated to (a) so that the first direction of orientation of (b) is substantially aligned with the first direction of orientation of (a).

2. A film substrate according to claim 1 wherein both (a) 15 and (b) further comprise a propylene copolymer skin on at least one side thereof.

3. A film substrate according to claim 2 wherein both (a) and (b) further comprise propylene copolymer skins on both the 20 inner and outer sides thereof.

4. A film substrate according to any of claims 1 to 3 wherein the propylene copolymer skins comprise from 10 to 20 wt% LDPE.

25

5. A film substrate according to any preceding claim wherein (b) is laminated to (a) by thermal lamination.

6. A film substrate according to any of claims 1 to 4 30 wherein the multilayer film substrate further comprises (c) a laminating adhesive resin between the inner sides of (a) and (b).

7. A film structure according to claim 6 wherein the 35 laminating adhesive resin comprises low density polyethylene (LDPE) and/or linear low density polyethylene (LLDPE).

- 17 -

8. A film substrate according to claim 6 or 7 wherein a primer is provided between the propylene copolymer skins on the inner sides of (a) and (b) and the laminating adhesive resin.

5

9. A film substrate according to claim 6, 7 or 8 wherein (c) is of ABA construction, comprising a core layer B of LLDPE and layers A of LDPE.

10 10. A film structure according to any preceding claim 6, 7, 8 or 9 wherein the laminating adhesive resin comprises an orientable component which is oriented in the machine direction, resulting in said resin being oriented substantially perpendicular to said first and second layers.

15

11. A film substrate according to any preceding claim wherein the first direction is transverse direction (TD) and the second direction is machine direction (MD).

20

12. A film substrate according to any preceding claim further comprising (d) a security device between (a) and (b).

25

13. A film substrate according to claim 12 wherein (d) is provided between the primer coatings on the inner sides of (a) and (b).

30

14. A film substrate according to claim 13 wherein (d) comprises at least one of an optically variable device (OVD), a magnetic device, an electronic device, or a rare earth element-containing device.

35

15. A film substrate according to claim 14 wherein (d) comprises an OVD which comprises at least one of a diffraction grating, a liquid crystal, a moire pattern produced by cross-gratings with or without superimposed, refractive, lenticular and transparent grids, a spaced partially-reflective, and partially transparent, coating yielding variable interference

- 18 -

patterns, a bi-refringent or polarising layer or a zone-plate.

16. A film substrate according to claim 15 wherein the OVD comprises a reflecting diffraction grating comprising a 5 metallized thermoplastic film embossed with a diffraction pattern.

17. A film substrate according to any preceding claim wherein at least one outer side is externally coated with an 10 opacifying coating.

18. A film substrate according to claim 17 further comprising a clear window through the film substrate.

15 19. A film substrate according to claim 18 wherein the window is in register with an OVD thereby rendering it visible from either side of said substrate.

20. A film substrate according to any preceding claim in 20 the form of a security token, such as a bank note.

# INTERNATIONAL SEARCH REPORT

International application No.  
PCT/US95/08164

## A. CLASSIFICATION OF SUBJECT MATTER

IPC(6) : B32B 27/08

US CL : 428/500, 515, 516, 517, 523

According to International Patent Classification (IPC) or to both national classification and IPC

## B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

U.S. : 428/500, 515, 516, 517, 523

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched  
NONE

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

NONE

## C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	US, A, 4,943,780 (REDDING) 24 July 1990, see Abstract.	1-20

Further documents are listed in the continuation of Box C.

See patent family annex.

* Special categories of cited documents:	*T*	later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention
*A* document defining the general state of the art which is not considered to be of particular relevance	*X*	document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone
*E* earlier document published on or after the international filing date	*Y*	document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is taken alone
*L* document which may throw doubt on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)	*&*	document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art
*O* document referring to an oral disclosure, use, exhibition or other means		document members of the same patent family
*P* document published prior to the international filing date but later than the priority date claimed		

Date of the actual completion of the international search

09 SEPTEMBER 1995

Date of mailing of the international search report

24 OCT 1995

Name and mailing address of the ISA/US  
Commissioner of Patents and Trademarks  
Box PCT  
Washington, D.C. 20231

Faxsimile No. (703) 305-3230

Authorized officer

MARY CRITCHAR

Telephone No. (703) 308-2351

**THIS PAGE BLANK (USPTO)**